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Ab initio study of the quantum electronic coherences emerging from molecular attosecond ionization MARCO RUBERTI, Imperial College London — Here we present a new ab initio method for many-electron dynamics in polyatomic molecules, time-dependent B-spline Restricted Correlation Space-Algebraic Diagrammatic Construction (RCS-ADC)[M. Ruberti, Phys.Chem.Chem.Phys.21,17584(2019)]. RCS-ADC describes molecular ionization from first principles by combining the accurate description of electron correlation of quantum chemistry with the full account of the photoelectron continuum dynamics. We study the correlated many-electron dynamics upon attosecond ionization of pyrazine, providing a complete characterisation of the ionised state. We calculate the density matrix of the bipartite ion-photoelectron system, the ionic reduced density matrix (R-IDM), including description of correlated shakeup states, and the von Neumann entropy of the prepared entangled state. We study the ion-photoelectron interaction and unravel the mechanisms of onset and loss of quantum electronic coherences. Simulating the X-ray attosecond transient absorption measurement, by solving the von Neumann equations for a characterized R-IDM interacting with the X-ray probe field, allows us to unveil the mapping of the measurement results onto the non-zero R-IDM matrix elements. This study paves the way to the control of charged-directed photo-chemical reactivity.

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